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Measurement of TE-NORM Concentrations and Resulting Dose Assessment in Produced Water at Some Oil and Gas Sites

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ABSTRACT

Huge amounts of produced water that are TE-NORM-contaminated during the processes of extracting crude oil from oil&gas production fields with concentrations higher than the radiological reference levels provided by international organizations may expose the workers at these sites to potential radiation hazards. Therefore, assessing the worker doses at these sites have a great importance. This assessment is essential in determining the radiation hazards brought on by radiation exposure. The purpose of this study is to determine the activity concentration of TE-NORM contaminated produced water in different Egyptian oil&gas extraction sites, as well as using the estimated annual doses and radiation hazard indices to assess the radiation risks indices for the workers. The obtained data shows that the activity concentration of ^{238}U , ^{232}Th , and ^{40}K ranged from 4032 to 14567 Bq Kg⁻¹, 1185 to 6358 Bq Kg⁻¹ and 184 to 423 Bq Kg⁻¹ respectively. The calculated absorbed dose rate ranged from 2588 – 10587 nGy/h, and the calculated Annual Effective Dose Equivalent differ according to the TE-NORM activity concentrations and have the range of 3.17 to 12.99 mSv/y. It was found that the radiation hazard indices significantly more than the international values. Based on the obtained results, it could be concluded that discharging the contaminated produced water into the environment should be prohibited and using the recommended disposal methods to reduce as much as possible the exposure of radiation worker in oil & gas production fields.

INTRODUCTION

TE-NORM is an abbreviation for Technologically Enhanced Naturally Occurring Radioactive Material. It refers to any natural radioactive isotopes, which is present in the environment in which human activities have increasing its concentrations [1-4].

The use of recent techniques in oil & gas extraction sites has caused in the production of TE-NORM contaminated produced water as a by-products and wastes which may also caused from numerous industries such as uranium mining, phosphate production, coal ash, and metal-production processes [5-6]. U, Th and their relevant decay progenies is the major radionuclides in TE-NORM [6-7]. TE-NORM could accrue at different components along the oil and gas extraction process. Locations such as pipes,

pumps, separators, tanks and another tools of processing may be contaminated. TE-NORM can occur in form of scales, sludge, scraping, and produced waters. This may be a source of potential hazard of radiation to workers, general public, and the entire environment in case of definite measures are not recognized [8- 9]. Produced water contains formation water from the reservoir and/or condensed water (with gas production) [13]. Volumes of produced water differ significantly between installations and over the lifetime of the oil & gas fields, with a range of 2400–40 000 m³/d for oil production sites and 1.5–30 m³/d for gas production sites. Concentrations of up to a few hundred Becquerels per liter of ^{226}Ra , ^{228}Ra , ^{224}Ra and ^{210}Pb may present in the produced water [13].

TE-NORM may contain ^{226}Ra activity concentrations much higher than the exemption levels assigned by IAEA [10]. Less than 1 Bq/g for any radionuclide in the uranium decay chain or the thorium decay chain and less than 10 Bq/g for ^{40}K is the recommended exemption level for radionuclides of natural origin [14]. The first evaluations of occupational radiation exposure in the oil and gas industries were reported a few decades ago [11].

Activity levels of radiation depend on the formation stones that formed the reservoirs and the accompanying saline water, levels in the generated water may exceed several hundred Becquerels per liter [12-13]. Discharging this contaminated generated water into the environment might contaminate surface and ground water and expose both public and workers. [15].

Gamma radiation may cause radiation exposure for workers at oil&gas sites while performing their everyday tasks. This exposure to external gamma radiation is brought on by radionuclides that precipitate inside pipes and vessels as well as that which results from produced water that has been contaminated with TE-NORM. At oil and gas sites, the dosage rate may exceed tens of microSievert per hour [16].

This study aims to measure TE-NORM activity concentrations in the contaminated produced water in addition to, the hazard indices assessment of radiation for some Egyptian oil and gas sites workers. Produced water samples were collected from four sites at the eastern desert to calculate radium equivalent activity (Ra_{eq}), absorbed dose rate (D), annual effective dose rate (AEDE), internal hazard (H_{in}), external hazard (H_{ex}) and Gamma radiation representative level Index ($I\gamma$) for these sites personnel.

MATERIAL AND METHODS:

Collection of Samples:

The samples of TE-NORM contaminated produced water were collected from four oil and gas

production sites at the eastern desert, Egypt. Onshore production oilfield at the western bank of Suez Gulf, near to Hurghada city, about 450 kilometers from Cairo (Site 1), Onshore production oilfield at the western bank of Suez Gulf, near to Hurghada city, about 400 kilometers from Cairo (Site 2), Onshore production oilfield at the western bank of the Suez Gulf, near to Ras Ghareb city about 300 kilometers from Cairo (Site 3). Onshore production oilfield at the eastern bank of the Suez Gulf, near to Abu Rudeis city about 300 kilometers from Cairo (Site 4). Figure (1) shows sites location.

Three water samples collected from the sample point at the last stage of Oil-Water separation before water final disposal at each site. Two liters from each sample collected, acidified with 1 M HCl solution immediately to avoid the adsorption of radionuclides on the walls of the container, transferred to plastic bottles and transferred to laboratory for the radiometric measurement.

Samples preparation

Water samples volume reduced by evaporation at 60 °C to increase the activity concentration to be measurable. Two liters were reduced to one liter. An accurate 100 cm³ of each condensed water samples were putted in a PVC cylindrical radon-tight 100 cm³ Marinelli beaker, closed firmly using sealing tape in order to radon escape prevention, and kept for 28 days before the radioactivity analysis (to allow establishing the secular equilibrium between radium and its daughters).

Radioactivity measurements

The prepared samples were γ -assessed to define the specific activity of (^{226}Ra , ^{232}Th , and ^{40}K) using a shielded high-purity germanium (HPGe) detector with two inner concentric cylinders of lead, copper, and cadmium. A vertical Canberra N-type closed end-coaxial Canberra N-type HPGe detector (model GR4020) with about 40% relative photo peak efficiency and the resolution of this spectrometer



Fig (2): Sites location

was 2.0 keV energy resolution at 1.33 MeV photons of ^{60}Co was used. The spectra were analyzed using CANBERRA (Genie 2000) software for gamma acquisition and analysis.

According to the Genie- 2000 Spectroscopy Software manual, standard point sources is used to perform the regular efficiency calibration of the analyzer channels of the HPGe detector. After the measurement and subtraction of the background, the activity concentrations were calculated. The ^{226}Ra concentration were calculated by averaging over the measured activity for ^{214}Pb and ^{214}Bi .

- 1) ^{226}Ra activity was determined by measuring:
 - ^{214}Pb γ -rays; 295.1 (19.2%) and 352 (37.1%) keV.
 - ^{214}Bi γ -rays; 609.3 (46.1%) and 1120.3 (15%) keV.
- 2) ^{232}Th activity was determined by measuring:
 - ^{212}Pb γ -peaks of 238.6 (44.6%) keV.
 - ^{208}Tl γ -rays; 338.4 (12%), and 583.0 (30%) keV.

- 3) ^{40}K activity was Identified by measuring its 1460 (10.7%) keV γ -line.

In order to obtain a γ -spectrum with appropriate statistics, the counting period was around 60,000 seconds.

Radiological Effect Calculations

Absorbed dose rate (D)

The external radiation exposure from naturally occurring radionuclides can be determined terms of absorbed dose rate to assess radiological risk. The activity concentrations of the natural radionuclides ^{238}U , ^{232}Th , and ^{40}K are directly related with the absorbed dose rate in the air at 1 metre above the ground surface. Using the following formula, the contribution of terrestrial gamma radiation to absorbed doses in air is determined. [16-18]:

$$D (\text{nGy h}^{-1}) = 0.462 A_{\text{U}} + 0.604 A_{\text{Th}} + 0.042 A_{\text{K}} \quad (1)$$

where:

- D is the absorbed dose rate in nGy h^{-1} ,
- A_{U} , A_{Th} and A_{K} are the activity concentrations of ^{238}U , ^{232}Th and ^{40}K in Bq kg^{-1} respectively.

Annual effective dose equivalent (AEDE)

The conversion coefficient from the absorbed dose in the air to the effective dose and the outdoor occupancy factor can be used to estimate the annual effective dose equivalent from outdoor gamma radiation.

For the conversion coefficient from the absorbed dose in the air to the effective dose received by adults, a value of 0.7 Sv Gy^{-1} can be used, The UNSCEAR proposed outdoor occupancy factor is 0.2 assumes that workers spend roughly 8 working hours each day, depending on their living arrangements, and that the number of working days is set to be 50% of a year days. Using the following equation, the yearly effective dosage equivalent (mSv y^{-1}) was determined [1,16]:

$$\text{AEDE (mSv y}^{-1}\text{)} = D (\text{nGyh}^{-1}) \times 8760 \times C \times F \times 10^{-6} (\text{mSv nGy}^{-1}) \quad (2)$$

where

- D = the absorbed dose rate in nGy h^{-1} ,
- C = the occupancy factor,
- F = the conversion coefficient and,
- 8760 are the number of the year hours.

Radiation hazarded indices:

Radium equivalent radioactivity (Ra_{eq}):

The following equation is used to calculate the Radium equivalent radioactivity (Ra_{eq}) [1,16]:

$$\text{Ra}_{\text{eq}} = A_{\text{Ra}} + 1.43 A_{\text{Th}} + 0.077 A_{\text{K}} \quad (3)$$

where: A_{Ra} , A_{Th} and A_{K} are the activity concentration of ^{226}Ra , ^{232}Th and ^{40}K in Bq Kg^{-1} , respectively.

Gamma radiation level index (I_γ):

The following equation is used to calculate the gamma index (I_γ) as proposed by the European Commission [19].

$$I_\gamma = (A_{\text{U}}/300 + A_{\text{Th}}/200 + A_{\text{K}}/3000) \quad (4)$$

where, A_{U} , A_{Th} and A_{K} are the activity concentration of ^{238}U , ^{232}Th , and ^{40}K in Bq Kg^{-1} , respectively.

The external hazard index (H_{ex}):

The following equation is used to calculate the external hazard index (H_{ex}):

$$H_{\text{ex}} = (A_{\text{Ra}}/370 + A_{\text{Th}}/259 + A_{\text{K}}/4810) \leq 1 \quad (5)$$

Where A_{Ra} , A_{Th} and A_{K} are the specific activities of ^{226}Ra , ^{232}Th and ^{40}K in Bq Kg^{-1} , respectively [19].

The internal hazard index (H_{in}):

The following equation is used to calculate the internal hazard index (H_{in}):

$$H_{\text{in}} = (A_{\text{Ra}}/185 + A_{\text{Th}}/259 + A_{\text{K}}/4810) \leq 1 \quad (6)$$

where A_{Ra} , A_{Th} and A_{K} are the specific activities of ^{226}Ra , ^{232}Th and ^{40}K in Bq Kg^{-1} , respectively [20].

RESULTS AND DISCUSSION

Radioactivity measurements

Table 1 shows the radioactivity measurements of the contaminated produced water. These data show that, it is obvious that the specific activity varies greatly from location to location depending on how the source rock for the crude oil reservoirs was formed.

Table (1): Radioactivity measurements of the contaminated produced water

Site	Sample	Specific Radioactivity Bq Kg^{-1}		
		^{226}Ra	^{232}Th	^{40}K
Site 1	1	14486	6369	434
	2	14627	6356	425
	3	14588	6349	411
	Average	14567	6358	423.33
Site 2	1	10425	4820	240
	2	10532	4865	221
	3	10435	4853	235
	Average	10464	4846	232
Site 3	1	6732	2502	189
	2	6745	2592	180
	3	6740	2597	183
	Average	6739	2563.66	184
Site 4	1	4030	1191.00	222.40
	2	4045	1187.00	233.82
	3	4023	1177.00	220.45
	Average	4032.67	1185	225.56

For Sites 1, 2, 3, and 4, the average activity concentrations for ^{238}U were determined to be 14567, 10464, 6739, and 4032 Bq Kg⁻¹, respectively. For Sites 1, 2, 3, and 4, the average activity concentrations for ^{232}Th were determined to be 6358, 4846, 2563, and 1185 Bq Kg⁻¹, respectively. For Sites 1, 2, 3, and 4, the corresponding average activity concentrations for ^{40}K were found to be 423, 232, 184, and 225 Bq Kg⁻¹.

The activity concentrations levels of the Uranium and Thorium series of the contaminated produced water samples varies from site to site according to the oil reservoirs source rocks. Therefore, Sites 1, 2, 3, and 4 exhibited different activities despite being nearby (in the eastern desert).

Radiological Effect Calculations

Absorbed dose rate (D)

Table 2 shows the calculated absorbed dose rate. The average calculated absorbed dose rate ranged from 2588 to 10578 nGy/h depending on the activity concentration of TE-NORM contamination. Site 1 exhibited the greatest calculated absorbed dose rate of approximately 10587 nGy/h with the highest concentration of NORM activity. Site 4 exhibited the lowest calculated absorbed dose rate of around 2588 nGy/h with the lowest activity concentration of TE-NORM.

Table (2): D and AEDE

Site	Sample	specific radioactivity Bq Kg ⁻¹			Absorbed Dose Rate D (nGy/h)	Annual Effective Dose Equivalent mSv/y
		^{238}U	^{232}Th	^{40}K		
Site 1	1	14486	6369	434	10557.64	12.95
	2	14627	6356	425	10614.55	13.02
	3	14588	6349	411	10591.71	12.99
	Average	14567	6358	423.33	10587.97	12.99
Site 2	1	10425	4820	240	7737.71	9.49
	2	10532	4865	221	7813.53	9.58
	3	10435	4853	235	7762.05	9.52
	Average	10464	4846	232	7771.10	9.53
Site 3	1	6732	2502	189	4629.33	5.68
	2	6745	2592	180	4689.32	5.75
	3	6740	2597	183	4690.15	5.75
	Average	6739	2563.67	184	4669.60	5.73
Site 4	1	4030	1191.00	222.40	2590.56	3.18
	2	4045	1187.00	233.82	2595.56	3.18
	3	4023	1177.00	220.45	2578.79	3.16
	Average	4032.67	1185	225.56	2588.31	3.17

Annual effective dose equivalent (AEDE)

The calculated values of the annual effective dose equivalent showed in table (2). The calculated Annual Effective Dose Equivalent for Site 1, which had the highest TE-NORM activity concentration, was found to be about 12.99 mSv/y. While the calculated Annual Effective Dose Equivalent for Site 4, which had the lowest TE-NORM activity concentration, was found to be about 3.17 mSv/y.

According to the level of TE-NORM contamination activity, the average Annual Effective Dose Equivalent for Sites 1, 2, 3, and 4 has varied from 3.17 to 12.99 mSv/y. AEDE is significantly greater than the corresponding global value of 0.50 mSv/y. [1, 16, 21].

Relation between the activity concentrations of the TE-NORM contamination and the AEDE of workers showed in figure (2).

The values of the AEDE for employees working in TE-NORM-contaminated areas at oil and gas sites demonstrate that the external effective dose is affected by the levels of activity of TE-NORM contamination in those areas (the higher the activity concentration, the higher the AEDE).

Radiation hazarded indices

In oil&gas extraction fields, the natural activity concentration of TE-NORM contamination typically

resulted from the contents of ^{226}Ra , ^{232}Th , and ^{40}K . ^{226}Ra and its daughter products produce 98.5% of the radiological hazards of the ^{238}U series, so the contribution from the ^{238}U has been replaced with the ^{226}Ra decay products. The gamma-ray radiation hazards due to the specified radionuclides has been assessed using different indices. These indices like Radium equivalent radioactivity (Ra_{eq}), Gamma radiation level index (I_γ) and the external and internal hazard index (H_{ex} , H_{in}) [16, 23, 24].

Radium equivalent radioactivity (Ra_{eq}):

Radium equivalent radioactivity (Ra_{eq}) is the first index that has been established to express the specific activities of ^{226}Ra , ^{232}Th and ^{40}K by a single quantity, which takes into account the health risks effects that produced from ^{238}U , ^{232}Th , and ^{40}K activity concentrations [19-20]. The external gamma dose and internal dose due to radon and its daughters are related to Ra_{eq} . For safe use, Ra_{eq} levels in natural radioactivity must be no higher than 370 Bq Kg^{-1} [17, 26].

Table (3) showing the calculated values of the radium equivalent radioactivity (Ra_{eq}). The data shows that, for sites 1, 2, 3, and 4 the radium equivalent activities ranged from 5744 Bq Kg^{-1} to 23691 Bq Kg^{-1} with an average 14316 Bq Kg^{-1} , which is more than 370 Bq Kg^{-1} that is the international permissible limit for safe handling usage [1,16].

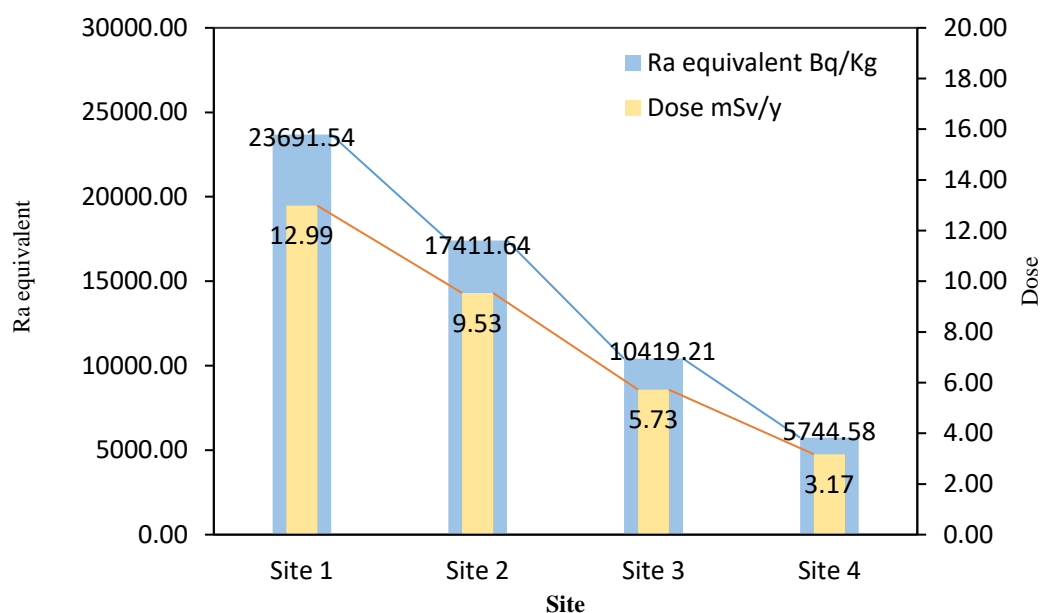


Fig (2): Relation between the activity concentrations of the TE-NORM contamination and the workers AEDE

Table (3): Radiation Hazardred Indices

Site	Sample No.	Radium Equivalent R_{aeq} (Bq Kg ⁻¹)	Gamma Index I_γ	Internal Hazard Index H_{in}	External Hazard Index H_{ex}
Site 1	1	23627.09	102.98	63.83	80.28
	2	23748.81	103.69	64.16	80.68
	3	23698.72	103.45	64.03	80.51
	Average	23691.54	103.38	64.01	80.49
Site 2	1	17336.08	75.01	46.84	58.93
	2	17505.97	75.76	47.29	59.51
	3	17392.89	75.19	46.99	59.13
	Average	17411.64	75.32	47.04	59.19
Site 3	1	10324.41	46.09	27.89	35.01
	2	10465.42	46.50	28.27	35.50
	3	10467.80	46.50	28.28	35.51
	Average	10419.21	46.36	28.15	35.34
Site 4	1	5750.25	26.43	15.54	19.46
	2	5760.41	26.50	15.56	19.50
	3	5723.08	26.34	15.46	19.37
	Average	5744.58	26.42	15.52	19.44
Average	14316.74	62.87	38.68	48.62	

Gamma radiation level index (I_γ):

Gamma radiation level index (I_γ) was suggested by the European commission to assess the hazardous level of radionuclides in the human body when exposed to external annual effective doses of gamma radiations from radionuclides decay. This index is very important in monitoring radiation inside human body and for quality control of gamma radiation annual effective doses to ensure that such radiation does no higher than the worldwide permissible high dose values [18, 25].

0.3 mSv/y is the exemption dose criterion that corresponds to $I_\gamma \leq 0.5$, whereas 1 mSv/y the dose criterion corresponds to $I_\gamma \leq 1$ [20]. On other hand, the corresponding values of I_γ for superficial and other materials should be between 2 and 6.

Table (3) shows the calculated I_γ . The data shows that, for sites 1, 2, 3, and 4 the gamma index (I_γ) ranged from 26 to 103 which is considerably greater than the assigned levels recommended by European commission for safe handling use.

External and internal hazard index (H_{ex} , H_{in}):

Radium expression can provide the external hazard index. To keep the hazards of radiation insignificant this index value must be less than unity; in other words, the maximum annual external radiation dose due to NORM radioactivity is 1.0 mSv. The internal exposure to carcinogenic radon and its short-lived daughters can be indicated by the internal hazard index (H_{in}) [20].

Table (3) shows the data obtained of the calculated external and internal hazard indexes. The obtained data shows that, for sites 1, 2, 3, and 4 the external hazard index (H_{ex}) ranged from 19 to 80 with an average 48.6 which is considerably greater than the international assigned values ($H_{ex} < 1$).

For sites 1, 2, 3, and 4 the internal hazard index (H_{in}) ranged from 15 to 64 with an average 38.7 which is considerably greater than the international assigned values ($H_{in} < 1$).

The values of the absorbed dose rate, annual effective dose equivalent, and radiation hazard indices for all investigated sites 1, 2, 3, and 4 are clearly greater than international levels, as can be seen from the data above. This largely depends on the levels of NORM activity that are being released into the environment through the produced water (the higher the activity the higher the absorbed dose rate, annually effective dose equivalent, and the radiation hazard indices).

CONCLUSIONS

The activity concentrations of ^{238}U , ^{232}Th and ^{40}K in the produced water samples have been obtained using gamma-ray spectrometry. The activity concentrations of ^{238}U , ^{232}Th and ^{40}K exceeds the exemption levels of the international regulations. The values of the calculated absorbed dose rate, calculated Annual Effective Dose Equivalent, radiation hazard indices was calculated and found to be much higher than the international values.

TE-NORM contaminated produced water from oil and gas industry has been generated radiation exposure levels, which require consideration and continuous monitoring during routine work in this industry. According to ALARA principles, it is important to restrict the areas that can be contaminated and rotate workers' jobs. It is also important to reduce as much as possible the amount of time spent in contaminated areas. In order to limit worker exposure to radiation as much as possible, regulations have to mandate oil and gas sites to use the allowed disposal methods and prevent from releasing contaminated produced water into the environment.

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